

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Patent Application of:  
Liliana Bagala Rampazzo et al.

Application No.: 10/523,101

Confirmation No.: 8413

Filed: February 1, 2005

Art Unit: 1709

For: SPIROBIFLUORENE DERIVATIVES, THEIR  
PREPARATION AND USES THEREOF

Examiner: M. E. Nelson

**1.132 DECLARATION**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Dear Sir:

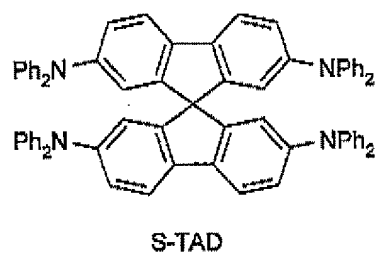
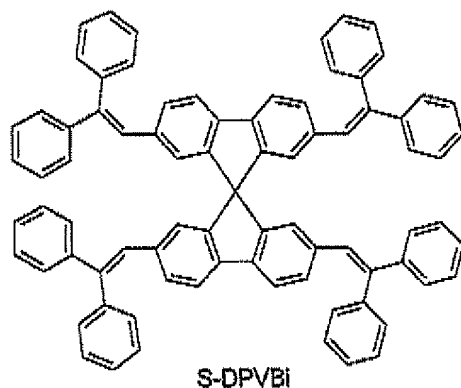
1. I, Dr. Philipp Stoessel am a citizen of the Federal Republic of Germany and reside at Sophienstrasse 30, 60487 Frankfurt, Germany, hereby declare and say as follows:
2. I am a fully trained chemist, having studied chemistry at the University of Tuebingen, Baden-Wuerttemberg, Germany.  
I am well acquainted with technical English.
3. Work experience:  
1986 – 1996: Studies in chemistry at the University of Tuebingen, Germany  
1996 – 1998: Postdoctoral research in chemistry sponsored by the  
Alexander von Humboldt Foundation with Dr. J. R. Norton at the  
Colorado State University, Fort Collins, CO and at the Columbia  
University New York, NY.  
1998 – 1999: Material Scientist and Synthetic Chemist at the Institut fuer Neue  
Materialien, Saarbruecken, Saarland, Germany

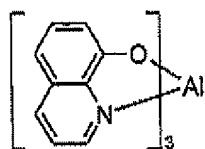
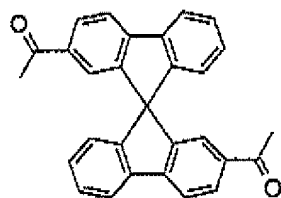
1999 – 2005: Material Scientist and Synthetic Chemist at COVION Organic  
Semiconductors GmbH, Frankfurt, Hessen, Germany

2005 – today: Material Scientist and Synthetic Chemist at the Merck Organic  
Materials GmbH, Frankfurt, Hessen, Germany

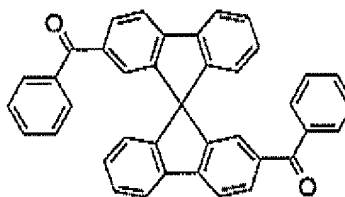
4. In the field of OLED, I am inventor up to now of at least 73 World wide patent and patent publications which include at least 70 U.S. patents and US patent publications and author of more than 10 publications and lectures.
5. In view of my qualifications as outlined above, I consider myself to be an expert and to be skilled in the spirobifluorene (SBF) compounds and organic light emitting diode (OLED) field.
6. I have read and reviewed Lupo, U.S. patent 5,840,217 ("Lupo").
7. I have read and reviewed this application, U.S. Application No. 10/523,101 (" '101 application").
8. I had the following experiment conducted under my supervision.
9. OLEDs are produced by a general process as described in WO 04/058911, which is adapted in individual cases to the particular circumstances (for example layer-thickness variation in order to achieve optimum efficiency).
10. The results of various OLEDs are presented in the following examples. The basic structure, the materials and layer thicknesses used, apart from the electron-transport layer, are identical for better comparability. OLEDs having the following structure are produced analogously to the above-mentioned general process:
11. Hole-injection layer (HIL) 60 nm PEDOT (spin-coated from water; purchased from H. C. Starck; poly(3,4-ethylenedioxy-2,5-thiophene)
12. Hole-transport layer (HTL) 20 nm NaphDATA (vapour-deposited; purchased from SynTec; 4,4',4''-tris(N-1-naphthyl-N-phenylamino)-triphenylamine
13. Hole-transport layer (HTL) 20 nm S-TAD (vapour-deposited; prepared in accordance with WO 99/12888; 2,2',7,7'-tetrakis(diphenylamino)spiro-9,9'-bifluorene)
14. Emission layer (EML) 30 nm spiro-DPVBi (prepared in accordance with WO 02/10093, 2,2',7,7'-tetrakis(2,2'-diphenylvinyl)spiro-9,9'-bifluorene) doped with 1% of S-TAD (2,2',7,7'-tetrakis(diphenylamino)spiro-9,9'-bifluorene)

15. Electron conductor (ETL) 20 nm – 40 nm (precise structure see examples in Table 1) (vapour-deposited:  $\text{AlQ}_3$  purchased from SynTec; tris(quinolinato)aluminium(III) or electron-transport materials E1 or E2 as shown below.
16. Ba/Al (cathode) 3 nm Ba, 150 nm Al on top.
17. For the characterization, the electroluminescence spectra, the efficiency (measured in  $\text{cd/A}$ ), the power efficiency (measured in  $\text{lm/W}$ ) as a function of the brightness, calculated from current-voltage-brightness characteristic lines (IUL characteristic lines), and the lifetime are determined. The lifetime is defined as the time after which the initial brightness of the OLED has dropped to half at a constant current density of  $10 \text{ mA/cm}^2$ . For the electron-transport layer, the layer thickness was optimized separately for each material. For better comparison, however, the greater  $\text{AlQ}_3$  layer thicknesses, which are directly comparable with the layer thicknesses of E1 and E2, are also shown.
18. Table 1 shows the results of some examples, with the composition of the ETL including the layer thicknesses also being shown in each case. As electron-transport material, the ETLs comprise compounds E1 or E2. The comparative examples used are OLEDs which comprise  $\text{AlQ}_3$  as electron conductor in accordance with the prior art.
19. For better clarity, the structural formulae of the substances used are shown below:



AlQ<sub>3</sub>

E1



E2

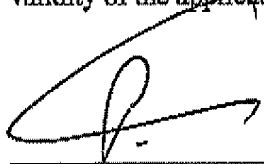
20. Table 1:

Example	ETL	Max. efficiency (cd/A)	Power efficiency @ 100 cd/m <sup>2</sup> (lm/W)	CIE (x, y)	Lifetime (h) at 10 mA/cm <sup>2</sup>
Example 1 (comparison)	AlQ <sub>3</sub> (20 nm)	4.2	1.8	0.17 / 0.23	700
Example 2 (comparison)	AlQ <sub>3</sub> (40 nm)	3.5	1.7	0.17 / 0.30	650
Example 3	E1 (40 nm)	4.5	2.3	0.17 / 0.23	500
Example 4	E2 (40 nm)	4.8	2.6	0.17 / 0.22	1050

21. It has surprisingly been found that the use of the aroyl substituted spirobifluorene in an organic electroluminescent device gives unexpectedly better results than the use of the corresponding acetyl substituted spirobifluorene. The experimental results establish that the inventive compounds show both unexpectedly better

efficiency and better lifetime compared to the corresponding acetyl compound. In contrast, the results obtained with the acetyl compound are comparable to Alq, which is a standard electron transport material according to the state of the art. These results are unexpected results which are not obvious from the disclosure by Lupo.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

  
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Date